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# Concentrations of organophosphate flame retardants in dust from cars, homes, and offices: An international comparison

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## ABSTRACT

Concentrations of a number of organophosphate flame retardants (PFRs) were measured in floor dust collected from living rooms in Australia ( $n = 42$ ), Canada ( $n = 14$ ), Germany ( $n = 22$ ), and Kazakhstan ( $n = 9$ ); cars from Australia ( $n = 39$ ) and Germany ( $n = 19$ ); and offices from Germany ( $n = 25$ ) and Kazakhstan ( $n = 8$ ). PFR concentrations in these samples were compared with each other and with previously reported data for PFRs in dust from similar microenvironments in the UK. Our data reveal significant between-country differences in both absolute concentrations and the relative abundance of specific PFRs in each of the microenvironments studied. Most notably, concentrations of TCIPP in UK living room dust (median =  $21 \mu\text{g g}^{-1}$ ) exceeded significantly ( $p < 0.05$ ) those in all other countries studied here; a substantial number of car dust samples contained elevated concentrations of TDCIPP, and German samples generally contained lower levels of PFRs in all microenvironments studied. In addition, PFRs were determined in dust samples collected from living room couches in both Australia ( $n = 41$ ) and the UK ( $n = 10$ ). The elevated concentrations of TCIPP in UK living room dust are likely attributable to the favoured use of this PFR in UK couch foam. This is indicated by concentrations of TCIPP in UK couch dust (median =  $610 \mu\text{g g}^{-1}$ ) exceeding significantly those in Australian couch dust (median =  $2.9 \mu\text{g g}^{-1}$ ). Moreover, concentrations of TCIPP in UK couch dust originating from couches 15 years old or less, display a marked relationship with the age of the couch, with concentrations in such samples increasing significantly ( $p < 0.01$ ) with couch age.

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## 1. Introduction

Recent restrictions worldwide on the use of polybrominated diphenyl ethers (PBDEs), have led to increased use of alternative flame retardants, such as organophosphate flame retardants (PFRs). As PFRs are used as additive flame retardants (FRs), their transfer from products in which they are used into the environment is relatively facile, and their presence in indoor dust has been reported in a number of studies [1–6,14,15,17,19,20,24,26,27,29,30].

We reported recently on concentrations of PFRs in samples of floor dust from UK cars, school classrooms, homes, and offices [11].

The currently available data on the adverse health effects of PFRs were reviewed recently [31]. In summary, chlorinated alkyl phosphates such as tris(2-chloroethyl) phosphate (TCEP), tris(2-chloroisopropyl) phosphate (TCIPP), and tris(1,3-dichloro-2-propyl)phosphate (TDCIPP) are suspected carcinogens, with other effects such as reduced thyroid hormone levels [25], contact dermatitis [12], and neurotoxicity [16] also reported for TDCIPP. For the non-chlorinated PFRs, reported impacts include links with altered hormone levels and decreased semen quality for triphenyl phosphate (TPHP) [25]; neurotoxicity for tri-cresylphosphate (TMPP) [7]; haemolytic effects for 2-ethylhexyl diphenyl phosphate (EHDPP) [22]; and increased risk of mucosal symptoms of sick housing syndrome linked with higher indoor concentrations of tri-*n*-butyl phosphate (TNBP) [23].

While our UK study found no significant relationships between PFR concentrations in dust from cars, classrooms, homes, and

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offices and the presence of putative PFR sources in such UK microenvironments [11]; the same study did highlight elevated concentrations of TCIPP in house dust and suggested that this was likely attributable to extensive use of TCIPP in couch foam, as reported in the US [28]. This study explores this further, by comparing concentrations of TCIPP in Australian couch dust and from living rooms in which the couch was located; hypothesising that significantly elevated concentrations of TCIPP in couch compared to floor dust, combined with significant positive correlation between the two groups, would indicate couches to be a significant source. Moreover, while our earlier UK study [11] highlighted possible international differences in the absolute concentrations and relative abundance of individual PFRs in indoor dust; disparities between the sampling and analytical methodology employed by the various laboratories conducting studies in different countries, introduces some uncertainty. As a result, this study employs identical dust collection and analytical procedures to evaluate differences in concentrations of PFRs in samples of indoor dust taken from a variety of microenvironment categories in each of the following countries: Australia, Canada, Germany, and Kazakhstan. Concentrations reported in these samples are compared with those reported previously for the UK. To the best of our knowledge, these data are the first reported for Kazakhstan.

## 2. Materials and methods

### 2.1. Sampling

Samples of settled dust were collected at various points over the period 2011 to 2012 (except for Kazakhstani samples that were collected in 2009) using previously reported methods [21]. Samples were collected from: cars in Australia ( $n = 39$ ) and Germany ( $n = 19$ ), living rooms in Australia ( $n = 42$ ), Canada ( $n = 14$ ), Germany ( $n = 22$ ), and Kazakhstan ( $n = 9$ ); as well as offices from Germany ( $n = 25$ ) and Kazakhstan ( $n = 8$ ). We also collected couch dust samples from Australia ( $n = 41$ ) and the UK ( $n = 10$ ). Australian samples were collected predominantly from Brisbane and Sydney, Canadian from Toronto, German from several different cities, Kazakhstani from Almaty and Astana, while UK samples were obtained in the Birmingham area. For offices and living rooms, samples were obtained by vacuuming a set area of floor ( $1 \text{ m}^2$  if carpeted,  $4 \text{ m}^2$  if bare floor) for a set duration (1 min if carpeted, 4 min if bare floor). For cars, the seats and the dashboard area were sampled for 2 min, with couch dust collected by vacuuming the areas in contact with the sitter for 2 min. Dust was retained within a nylon “sock” ( $25 \mu\text{m}$  mesh size), placed in the vacuum cleaner furniture attachment. Following collection, samples were passed through a  $500 \mu\text{m}$  mesh sieve prior to analysis.

### 2.2. Analysis

Consistent with our previous study of PFRs in UK indoor dust, we measured concentrations of the following PFRs: TDCIPP, TCIPP, TPHP, TNBP, EHDPP, TCEP, and TMPP. An exception to this was for 12, 6, and 10 samples of German car, living room, and office dust respectively, for which data have been reported previously [10] but in which EHDPP was not measured. Concentrations were determined via GC-MS in accordance with methods reported previously [10,11]. Briefly, dust samples (50 mg, accurately weighed), were treated with 100 ng each of  $d_{15}$ -TPHP and  $d_{27}$ -TNBP as internal (or surrogate) standards, and extracted via vortexing, sonication, and centrifugation with three successive aliquots of hexane:acetone (3:1 v/v, 2 mL). The combined extracts were reduced using a gentle

stream of  $\text{N}_2$  to incipient dryness and reconstituted with 1 mL hexane prior to elution through a pasteur pipette containing 1 g Florisil. Following initial elution with hexane (8 mL, fraction not analysed), PFRs were eluted with ethyl acetate (10 mL). This second fraction was reduced to near dryness under a stream of  $\text{N}_2$  prior to reconstitution with 100  $\mu\text{L}$  of 1 ng/ $\mu\text{L}$  triamylphosphate (TAP) in iso-octane as recovery determination (or syringe) standard. Final sample extracts were analysed via GC-EIMS using an Agilent 5975C MSD fitted with a DB-5ms column (30 m, 0.25 mm id, 0.25  $\mu\text{m}$  film thickness). The GC temperature programme was 90  $^\circ\text{C}$ , hold for 1.25 min, ramp 10  $^\circ\text{C}/\text{min}$  to 170  $^\circ\text{C}$ , ramp 5  $^\circ\text{C}/\text{min}$  to 240  $^\circ\text{C}$ , hold for 10 min, ramp 20  $^\circ\text{C}/\text{min}$  to 310  $^\circ\text{C}$ , hold for 10 min. The mass spectrometer was operated in selected ion electron ionisation mode, with Table SD-1 listing the ions monitored for each targeted compound.

Purchased standards of TCIPP, TDCIPP and TMPP contained different isomers. While the commercial TCIPP mixture consists of 3 different isomers, the third eluting isomer has a markedly lower response than the others, and can only be seen at higher concentrations. Thus we report TCIPP levels here as a sum of the 1st two eluting isomers only (referred to as TCIPP 1 and TCIPP 2) [8,11]. Likewise, consistent with our UK study [11], concentrations of TDCIPP and TMPP in this study are reported as the sum of both and all four isomers respectively.

### 2.3. QA/QC

One aliquot of SRM2585 (NIST, organics in dust) was analysed with every batch of 10 dust samples. As the samples reported here are part of a larger PhD study, a total of 56 aliquots of SRM2585 were analysed. Table SD-2 illustrates the high reproducibility of our method with relative standard deviations ranging between 6.4% and 14% for individual PFRs. Neither certified nor indicative values for our target PFRs are reported by NIST. Nonetheless, Table SD-2 compares our data with the average  $\pm \sigma_n$  (consensus) values obtained for SRM2585 in an interlaboratory trial of PFR analysis in environmental samples [8]. The good agreement between our reported concentrations and those reported in the interlaboratory trial is evidence that our data are consistent with those published by other researchers.

One blank (comprising pre-baked  $\text{Na}_2\text{SO}_4$  treated as a dust sample) was analysed with every sample batch (thus every 6th sample was a blank), and a total of 107 blanks were analysed. Field blanks were also collected. These consisted of pre-baked  $\text{Na}_2\text{SO}_4$ , taken to the sampling location, spread on aluminium foil and vacuumed as a normal sample – i.e. 50 mg of  $\text{Na}_2\text{SO}_4$  was analysed as a surrogate for dust. Concentrations in a batch of samples were not corrected for those detected in blanks where the concentration of the target PFR in the blank from the same batch was less than 5% of the lowest concentration in that batch. Where the PFR concentration in the blank was between 5% and 20% of the concentration in samples from that batch, concentrations were corrected accordingly via subtraction of the blank concentration. If blank concentrations exceeded 20% of those in samples from the same batch, all samples in that batch were discarded and reanalysed. Concentrations of TNBP, EHDPP, TDCIPP and TMPP were below detection limits in all blank samples analysed. In contrast, (expressed as ng PFR per g  $\text{Na}_2\text{SO}_4$  “dust”) low levels of TCEP (median =  $0.023 \mu\text{g g}^{-1}$ ), TCIPP (median =  $0.03 \mu\text{g g}^{-1}$ ), and TPHP (median  $0.006 \mu\text{g g}^{-1}$ ) were detected in a small proportion of blanks. Where appropriate, correction for these blank levels was conducted.

### 3. Results and discussion

#### 3.1. PFRs in living room dust from Australia, Canada, Germany, and Kazakhstan

A statistical summary of the concentrations of PFRs in all samples of living room dust analysed in this study is provided as Table 1, alongside data from our previous study of UK living room dust for comparison. Concentrations of PFRs in all individual samples analysed in this study are provided as Table SD-3. PFRs were detected in nearly all samples, with TCIPP the most abundant in Australia, Germany and the UK; while TPHP was the most abundant PFR in Canada and Kazakhstan. Using IBM SPSS Statistics for Mac (version 22.0.0.0), we applied ANOVA with Tukey post-hoc test to evaluate the hypothesis that significant differences exist between concentrations of individual PFRs in living room dust from the countries studied here. As visual inspection and a Kolmogorov-Smirnov test revealed the data were not normally distributed, concentrations were log-transformed prior to ANOVA. Table 2 summarises the significant differences revealed by this analysis. Two particularly salient features are that: (a) concentrations of TCIPP in UK living room floor dust exceed significantly those in the other four countries studied, and (b) that PFR concentrations in Germany are markedly lower than in any of the other countries.

#### 3.2. PFRs in dust from cars sampled in Australia and Germany

Table 3 summarises concentrations of target PFRs in samples of dust collected from cars in Australia and Germany, alongside our previously published data for UK cars [11]. Particular points of note here are that while TCIPP is predominant in cars sampled in Australia and the UK, it is another chlorinated PFR (TDCIPP) that predominates in cars sampled in Germany. Moreover, TDCIPP is present at substantial concentrations in Australian and UK car dust,

**Table 1**

Summary of concentrations ( $\mu\text{g g}^{-1}$ ) of PFRs in living room floor dust from Australia, Canada, Germany, Kazakhstan, and the UK.

PFR/Statistical Parameter	Australia	Canada	Germany	Kazakhstan	UK <sup>a</sup>
TnBP					
Minimum	<0.03	<0.03	<0.03	<0.03	<0.03
Median	0.06	0.13	<0.03	0.11	<0.03
Maximum	8.4	1.2	0.25	0.23	0.09
TCEP					
Minimum	<0.06	0.19	<0.06	0.62	<0.06
Median	0.60	0.69	0.21	1.4	0.81
Maximum	24	37	5.7	6.8	28
TCIPP					
Minimum	0.24	0.12	0.33	0.43	3.7
Median	1.8	1.2	1.0	1.0	21
Maximum	24	37	5.7	6.8	100
TPHP					
Minimum	0.24	0.02	0.07	1.2	0.49
Median	1.2	1.6	0.23	3.8	3.3
Maximum	31	37	18	9.2	110
EHDPP					
Minimum	<0.01	0.01	<0.01	0.06	0.18
Median	0.38	0.39	0.14	0.27	1.6
Maximum	5.1	0.73	0.56	1.2	130
TDCIPP					
Minimum	<0.03	0.03	<0.03	<0.03	0.06
Median	0.32	1.1	0.08	0.11	0.71
Maximum	11	3.2	14	2.0	14
TMPP					
Minimum	<0.01	<0.01	<0.01	<0.01	<0.01
Median	0.04	<0.01	0.14	<0.01	0.02
Maximum	3.0	0.67	1.3	1.1	14

<sup>a</sup> Reported in Ref. [11].

**Table 2**

Statistically significant ( $p < 0.05$ ) differences in concentrations of PFRs in living room floor dust from Australia, Canada, Germany, Kazakhstan, and the UK.

PFR	Significant difference
TnBP	Australia, Kazakhstan > UK
TCEP	Australia, Canada, Kazakhstan, UK > Germany
TCIPP	UK > Australia, Canada, Germany, Kazakhstan
TPHP	Canada, Australia, Kazakhstan, UK > Germany
TDCIPP	Australia, Canada, UK > Germany

**Table 3**

Summary of Concentrations ( $\mu\text{g g}^{-1}$ ) of PFRs in Car Dust from Australia, Germany, and the UK.

PFR/Statistical Parameter	Australia	Germany	UK <sup>a</sup>
TnBP			
Minimum	<0.03	<0.03	<0.03
Median	0.11	<0.03	<0.03
Maximum	8.4	0.63	1.2
TCEP			
Minimum	<0.06	<0.06	<0.06
Median	2.0	0.40	1.2
Maximum	62	5.1	8.7
TCIPP			
Minimum	0.31	0.29	2.4
Median	24	2.9	53
Maximum	310	100	370
TPHP			
Minimum	0.33	0.33	0.27
Median	3.7	1.8	3.3
Maximum	85	11	170
EHDPP			
Minimum	0.18	0.01	0.29
Median	0.63	1.2	2.2
Maximum	4.9	1.9	11
TDCIPP			
Minimum	0.06	<0.03	0.11
Median	2.3	4.1	31
Maximum	730	620	740
TMPP			
Minimum	<0.01	<0.01	<0.01
Median	0.31	0.86	0.59
Maximum	240	150	5.6

<sup>a</sup> Reported in Ref. [11].

and the maximum concentrations of any PFR in any microenvironment in this study are of TDCIPP in cars (730, 620, and 740  $\mu\text{g g}^{-1}$  in cars sampled in Australia, Germany, and the UK respectively). These higher concentrations of TDCIPP in car dust are consistent with other studies [9,13] and reports that TDCIPP is used only in applications requiring an especially high degree of flame retardancy due to its higher cost compared to TCIPP [18]. One such application is in polyurethane foam used in cars. As for living room dust we examined our data for significant differences in concentrations of target PFRs in cars from different countries, by subjecting log-transformed concentrations to ANOVA, and the significant differences detected by this analysis are shown in Table 4. The principal features are that concentrations of TCIPP, EHDPP, and TDCIPP in UK car dust exceed significantly those in cars

**Table 4**

Statistically significant ( $p < 0.05$ ) differences in concentrations of PFRs in car dust from Australia, Germany, and the UK.

PFR	Significant difference
TnBP	Australia > UK
TCIPP	Australia, UK > Germany
EHDPP	UK > Australia
TDCIPP	UK > Australia, Germany

from at least one of the other two countries studied. However, TnBP is rarely detected in UK cars and concentrations of this PFR in Australian vehicles significantly exceed those in the UK.

We also examined our data to check for any relationship between vehicle age and the concentrations of each target PFR. No significant relationships were detected, with one exception; that concentrations of EHDPP in cars sampled in Australia displayed a significant negative correlation ( $p = 0.03$ ) with vehicle age. This implies that concentrations of EHDPP are higher in newer vehicles. We also checked for any systematic influence of vehicle manufacturer on PFR concentrations and found none. As an illustration, while the concentration of TDCIPP in one Toyota Corolla made in 1997 was  $740 \mu\text{g g}^{-1}$ , in another 1997 Corolla, the concentration of the same PFR was  $0.12 \mu\text{g g}^{-1}$ . While our database is small, and a larger study may lead to different conclusions, this wide disparity in TDCIPP concentrations between these two identical car models manufactured in the same year, suggests that the factors influencing concentrations of PFRs in car dust are complex and multifactorial, and likely depend not only on the year and country of manufacture, but on additional factors such as foam cushions, car seats etc. introduced by the car owner.

### 3.3. PFRs in dust from offices in Australia, Germany, and Kazakhstan

Concentrations of target PFRs detected in dust taken from offices in Germany and Kazakhstan, alongside those in our earlier study of UK offices are summarised in Table 5. Of particular note, while TCIPP is the predominant PFR in German and UK office dust, TPHP is the major PFR in offices in Kazakhstan. ANOVA of log-transformed concentrations was performed to evaluate the existence of any significant differences in concentrations of target PFRs in office dust from the different countries studied. The significant differences revealed by this analysis are shown in Table 6, revealing concentrations of TCIPP, EHDPP, and TDCIPP in UK offices to significantly

**Table 5**  
Summary of Concentrations ( $\mu\text{g g}^{-1}$ ) of PFRs in Office Dust from Germany, Kazakhstan, and the UK.

PFR/Statistical Parameter	Germany	Kazakhstan	UK <sup>a</sup>
TnBP			
Minimum	<0.03	<0.03	<0.03
Median	0.17	0.07	<0.03
Maximum	0.76	0.48	1.3
TCEP			
Minimum	<0.06	0.95	<0.06
Median	0.13	2.5	0.87
Maximum	12	5.8	160
TCIPP			
Minimum	0.18	0.87	3.6
Median	1.6	2.2	33
Maximum	13	100	230
TPHP			
Minimum	0.20	0.39	0.56
Median	1.5	5.3	4.3
Maximum	8.8	48	50
EHDPP			
Minimum	0.13	0.08	0.15
Median	0.36	0.26	5.3
Maximum	3.8	0.57	81
TDCIPP			
Minimum	<0.03	<0.03	<0.03
Median	0.14	0.91	0.48
Maximum	2.2	4.0	51
TMPP			
Minimum	<0.01	0.01	<0.01
Median	<0.01	0.38	<0.01
Maximum	1.9	10	5.3

<sup>a</sup> Reported in Ref. [11].

**Table 6**

Statistically significant ( $p < 0.05$ ) differences in concentrations of PFRs in office floor dust from Germany, Kazakhstan, and the UK.

PFR	Significant difference
TCIPP	UK > Germany, Kazakhstan
EHDPP	UK > Germany, Kazakhstan
TDCIPP	UK > Germany

exceed those in Germany in all cases and Kazakhstan for TCIPP and EHDPP. While not significant, it is noteworthy that median concentrations of TCEP, TPHP, TDCIPP, and TMPP are all highest in Kazakhstani office dust.

### 3.4. Patterns of PFR contamination in dust from different countries and microenvironments

In addition to differences in absolute concentrations of target PFRs in dust from various microenvironments in different countries, we also examined our data for between-country variations in the relative abundance of individual PFRs. To do so, we first normalised concentrations of each individual PFR to  $\Sigma\text{PFRs}$  in each sample. We then subjected these normalised data to principal component analysis using SPSS. Figs. 1–3 show plots of the first two principal components (PC1 and PC2) obtained for living rooms, car, and office dust respectively. Combined, these two PCs accounted for 43, 48, and 50% of the variance in the datasets for living room, car, and office dust respectively.

For living room dust samples (Fig. 1), PC1 scores (y axis) are driven primarily in a negative direction by high relative abundances of TCIPP and in a positive direction by high relative abundances of TPHP. For PC2 (x axis), high scores result from an elevated relative abundance of EHDPP, with high relative abundances of TDCIPP and TCEP yielding lower, more negative PC2 scores. Consequently, while Fig. 1 shows some overlap between samples from different countries, three fairly distinct clusters are evident. All but five of the UK samples display negative PC1 scores, with most falling within the cluster shaded green at a diagonal across the bottom two quadrants. This arises principally due to the relative abundances of TCIPP (high) and EHDPP (low) in most UK samples. Most Australian and Canadian samples fall within the blue shaded cluster with low scores for both PC1 (driven by relatively high abundances of TPHP) and (particularly) PC2 – the latter arising as a consequence of high relative abundances of TDCIPP. Finally, all the Kazakhstani samples are located in the top (purple shaded) cluster, all with positive PC1 (due to a predominance of TPHP) and (with one exception) negative PC2 scores – the latter due to a high relative abundance of TCEP.

Looking at the PC score plot for car dust samples (Fig. 2), it is apparent that with just 3 exceptions, all UK samples fall into the green shaded cluster spanning the bottom two quadrants due to their negative PC1 (y axis) scores. These negative PC1 scores result from the high relative abundance of both TCIPP and TDCIPP in UK car dust. Differentiation between UK samples occurs as a result of the relative abundance of these two chlorinated PFRs in individual samples; those in which TCIPP predominates having high PC2 (x axis) scores, while those dominated by TDCIPP having low PC2 scores. Samples from other countries are spread more evenly throughout component space, though one German and 12 Australian samples in which TCIPP is substantially elevated are grouped tightly together in the bottom right quadrant of the plot.

The PC score plot for office dust samples is shown in Fig. 3. Two clear clusters are evident. The red shaded cluster lying in the bottom left quadrant of component space encompasses the majority of the UK samples. This arises due to the negative scores for both PC1 (y axis) and PC2 due respectively to high relative abundances of



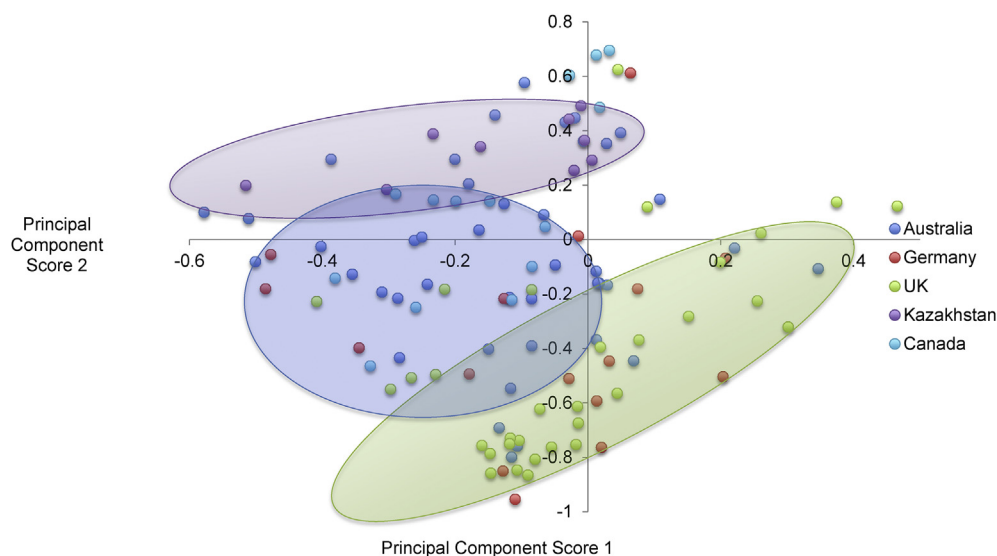


Fig. 1. Plot of scores for principal components 1 and 2 for living room floor dust samples.

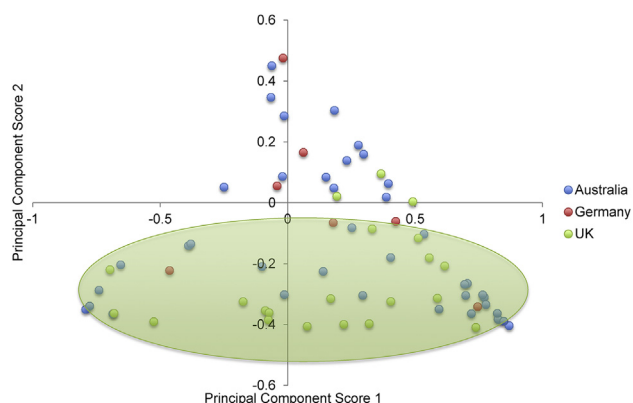


Fig. 2. Plot of scores for principal components 1 and 2 for car dust samples.

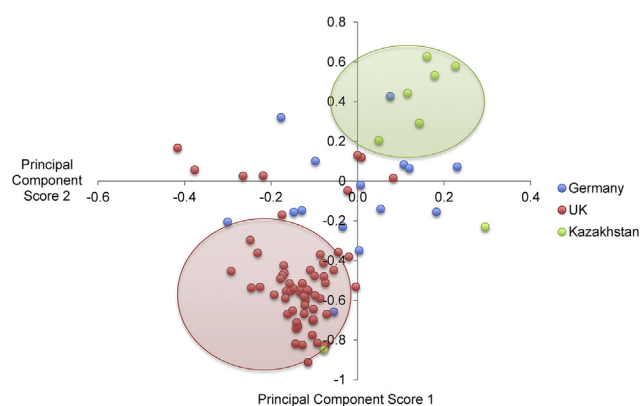


Fig. 3. Plot of scores for principal components 1 and 2 for office floor dust samples.

TCIPP and EHDPP in UK office dust. The other (green shaded) cluster that occupies the top right quadrant contains 6 out of the 8 Kazakhstani samples. This reflects the high proportions of TPHP (PC1) and TMPP (PC2) in these samples.

### 3.5. Do concentrations of TCIPP in Australian couch dust exceed those in matched floor dust samples?

Of the 41 couch dust samples collected in Australia, 40 corresponding floor dust samples (i.e. taken from the same room as the couch was located) were available. We therefore tested the hypothesis that concentrations of TCIPP in couch dust would exceed significantly those in corresponding floor dust samples. To do so, we conducted a paired t-test comparison of log-transformed concentrations in both datasets. This analysis revealed concentrations of TCIPP in couch dust (average =  $29 \mu\text{g g}^{-1}$ ) to exceed significantly ( $p = 0.014$ ), those in corresponding floor dust samples (average =  $4.1 \mu\text{g g}^{-1}$ ). This is consistent with the hypothesis that couches are a net source of TCIPP in Australian homes. However, this is contradicted by the absence of significant correlation ( $p > 0.1$ ) between log-transformed TCIPP concentrations in dust from couches and the corresponding living room floors. This latter observation indicates that couches are not the only source of TCIPP in Australian living rooms.

### 3.6. Do TCIPP concentrations in UK couch dust exceed significantly those in Australian couch dust?

A t-test comparison of log-transformed concentrations of TCIPP in Australian and UK couch dust reveals concentrations in UK couch dust (median =  $610 \mu\text{g g}^{-1}$ ) to exceed significantly those in Australian couch dust (median =  $2.9 \mu\text{g g}^{-1}$ ). This is consistent with the significantly higher TCIPP concentrations in UK compared to Australian living room dust (Table 2).

### 3.7. Influence of couch age on concentrations of TCIPP in couch dust

No discernible relationship was evident between couch age and concentrations of TCIPP in Australian couch dust. In contrast, the influence of couch age on concentrations of TCIPP in UK couch dust was examined by plotting TCIPP concentration against couch age for the 9 out of 11 couches for which information on the couch age was available (Fig. 4). When all data were plotted, no significant relationship was detected. However, one couch was antique and conservatively estimated by its owner to be around 50 years old. When this sample was removed as an outlier, a significant positive

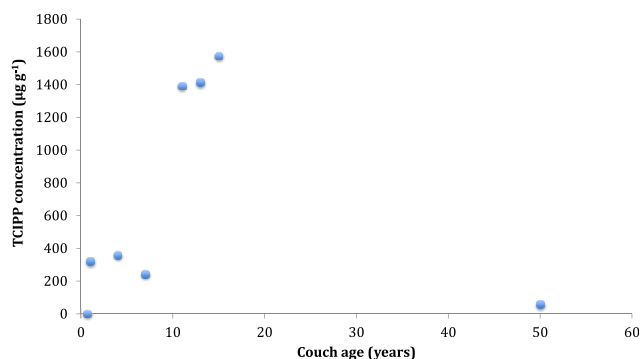


Fig. 4. Plot of concentration of TCIPP in UK couch dust Against couch age.

relationship ( $R = 0.93$ ,  $p < 0.01$ ) was evident between couch age for couches  $\leq 15$  years old and the concentration of TCIPP in the corresponding floor dust sample. Closer inspection of this small UK dataset, reveals the highest TCIPP concentrations ( $1400$ – $1600 \mu\text{g g}^{-1}$ ) to be in dust sampled from the 3 couches that were purchased between 11 and 15 years prior to dust sample collection. Concentrations of TCIPP in samples from couches sampled between 1 and 7 years were much lower ( $240$ – $360 \mu\text{g g}^{-1}$ ), with that in dust sampled from a couch purchased just 3 months hitherto only  $1.81 \mu\text{g g}^{-1}$  TCIPP. While more data are required to confirm these findings, this suggests that while couches are a substantial source of TCIPP in UK homes, use of this PFR in UK couches has declined substantially from a decade or so ago. While this will likely lead to a reduction in TCIPP concentrations in UK homes, it suggests that substantial quantities of soft furnishings containing TCIPP will enter the UK waste stream in coming years as unwanted furniture is discarded.

### 3.8. Assessment of human health risk arising from exposure to PFRs via dust ingestion

While exposure and risk assessment was not a primary objective of this study, we have previously done so for the UK population based on the same dust samples mentioned for comparison purposes in this paper [11]. In our earlier paper, we noted that the highest risk was for our high-end exposure estimate of toddler exposure to TCIPP, which based on UK data was  $\sim 5$  times lower than a health based limit value (HBLV) for this PFR [26]. Our UK exposure estimates for other PFRs were all at least 90 times lower than the corresponding HBLV. As this current study shows UK dust to contain the highest concentrations of TCIPP and most other PFRs, we conclude that exposure via dust ingestion in the other countries studied here will also not exceed existing HBLVs. It should be noted however, that current HBLVs have no legislative standing and that future advances in toxicological understanding may reduce these apparent margins of safety. Furthermore, consideration of other exposure pathways such as diet, inhalation, and dermal uptake may narrow erode further the gap between HBLVs and exposure.

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### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.emcon.2016.05.002>.

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